Universality of the Tangential Shape Exponent at the Facet Edge of a Crystal

Yasuhiro Akutsu

Department of Physics, Graduate School of Science, Osaka University, Toyonaka 560, Osaka, Japan

Noriko Akutsu

Faculty of Engineering, Osaka Electro-Communication University, Neyagawa 572, Osaka, Japan

Takao Yamamoto

Department of Physics, Faculty of Engineering, Gunma University, Kiryu 376, Gunma, Japan
(March 13, 1998)

Below the roughening temperature, the equilibrium crystal shape (ECS) is composed of both facets and a smoothly curved surface. As for the "normal" profile (perpendicular to the facet contour), the ECS has the exponent 3/2 which is characteristic of systems in the Gruber-Mullins-Pokrovsky-Talapov universality class. Quite recently, it was pointed out that the ECS have a "new" exponent 3 for "tangential" profile. We first show that this behavior is universal because it is a direct consequence of the well-established universal form of the vicinal-surface free energy (p: surface gradient): $f(p) = f(0) + \gamma p + Bp^3 + O(p^4)$. Second, we give a universal relation between the amplitudes of the tangential and the normal profiles, in close connection with the universal Gaussian curvature jump at the facet edge in systems with short-range inter-step interactions. Effects of the long-range interactions are briefly discussed.

PACS numbers: 05.70.Jk, 64.60.-i, 64.60.Fr, 68.35.Md, 68.35.-p, 82.65.Dp

A crystal surface with small average tilt relative to a crystal axis is called vicinal surface. Below the roughening temperature T_R the vicinal surface is well descibed by the terrace-step-kink (TSK) picture where the surface is made up of many "flat" terraces connected by steps. Since the step is a unidirectional linear object, the vicinal surface, as an assembly of steps, belongs to the Gruber-Mullins-Pokrovsky-Talapov (GMPT) universality class. [1] In this view, the vicinal-surface free energy $f(\rho)$ (per projected area) as a function of the step density ρ has the well-known form of the expansion, [1,2] $f(\rho) = f(0) + a_1\rho + a_3\rho^3 + O(\rho^4)$. This form of surface free energy leads to, via the Wulff construction, [3] the universal exponent 3/2 of the equilibrium crystal shape (ECS, for short) near the facet edge, $z \sim (\Delta x)^{3/2}$, where we have chosen the z axis to be facet normal, and Δx to be the "normal" distance from a facet edge. [4,5] Careful consideration of the crystal anisotropy gives us a universal relation [6] between the coefficients a_1 and a_3. This relation leads to the universal Gaussian curvature jump at the facet edge [6,7,8] whose physical origin is, therefore, quite different [9] from the universal curvature jump at T_R. [4]

Very recently, Dahmen *et al.* [10] pointed out that, if we approach the facet edge "tangentially" (i.e., along the tangential direction at the facet edge), ECS behaves as $z \sim (\Delta y)^3$ where Δy is the distance from the facet edge along the tangential direction. They drew this conclusion from an exact calculation (essentially equivalent to Ref.8) on the body-centered-cubic solid-on-solid model [11] (BCSOS model, for short). It is an interesting problem, then, to see whether this "new" exponent is universal or not. Also, relations, if any, to the above-listed universal behaviors are worth to be explored. To clarify these points is the aim of the present paper. As will be seen below, the tangential exponent 3 is a direct consequence of the GMPT-form of expansion of the surface free energy, hence, is universal. Further, the universal relation between the expansion coefficients leads also to a universal relation between the amplitudes of the normal and the tangential ECS profiles.

Consider a facet and its neighboring curved surface of the ECS with macroscopic size at a temperature T below T_R (of the facet, to be precise). We use the Cartesian coordinates (x, y, z) with z-direction chosen to be facet normal (downword, for convenience), which allows us to describe the ECS by an equation $z = z(x, y)$. The surface gradient at a position $(x, y, z(x, y))$ is conveniently expressed by the two-dimensional gradient vector $\boldsymbol{p} = (p_x, p_y)$ defined by

$$p_x = \frac{\partial z}{\partial x}, \quad p_y = \frac{\partial z}{\partial y}. \tag{1}$$

By $f(\boldsymbol{p})$ we denote the free energy per projected area [12] of the surface with fixed mean gradient $\boldsymbol{p}$. Introducing a "field" $\boldsymbol{\eta}$ conjugate to $\boldsymbol{p}$, we can perform the Legendre transformation $\boldsymbol{p} \to \boldsymbol{\eta}$, $f(\boldsymbol{p}) \to \tilde{f}(\boldsymbol{\eta})$, with

$$\tilde{f}(\boldsymbol{\eta}) = f(\boldsymbol{p}) - \boldsymbol{\eta} \cdot \boldsymbol{p}. \tag{2}$$

The Andreev free energy $\tilde{f}(\boldsymbol{\eta})$ directly gives us the ECS as [12]

$$z = -\frac{1}{\lambda}\tilde{f}(\lambda \boldsymbol{r}), \tag{3}$$

where $\boldsymbol{r} = (x, y)$ and λ is a scale factor. In what follows we consider "normalized" ECS by setting $\lambda = 1$ in (3). Then the field $\boldsymbol{\eta}$ is just the two-dimensional position vector of the ECS.

Properties of the ECS near the facet edge are governed by the small-$|\boldsymbol{p}|$ behavior of $f(\boldsymbol{p})$ which has the GMPT-type form of expansion. Properly taking account the crystal anisotropy, we have the expansion of the form,

$$f(\boldsymbol{p}) = f(0) + \gamma(\theta)|\boldsymbol{p}| + B(\theta)|\boldsymbol{p}|^3 + O(|\boldsymbol{p}|^4), \qquad (4)$$

where we have introduced the angle variable θ by

$$p_x = |\boldsymbol{p}|\cos\theta, \quad p_y = |\boldsymbol{p}|\sin\theta. \qquad (5)$$

Physically, $\gamma(\theta)$ is the step tension and θ the mean running direction angle of steps on the vicinal surface with gradient $\boldsymbol{p}$. Note that θ is also the direction angle of the tangential line of the facet contour (Fig.1). In systems with only short-range inter-step interactions, the coefficient $B(\theta)$ is always positive and is universally given by [6]

$$B(\theta) = \frac{\pi^2(k_{\mathrm{B}}T)^2}{6\tilde{\gamma}(\theta)}, \qquad (6)$$

where $\tilde{\gamma}(\theta) = \gamma(\theta) + \partial^2\gamma(\theta)/\partial\theta^2$ is the step stiffness. In association with the Legendre transformation, we have the following relation between the two-dimensional position vector $\boldsymbol{r} = (x, y)$ and the gradient $\boldsymbol{p}$:

$$\boldsymbol{r} = \frac{\partial f(\boldsymbol{p})}{\partial \boldsymbol{p}}. \qquad (7)$$

Near the facet edge in the curved region, we obtain from (4) (neglecting $O(|\boldsymbol{p}|^4)$ and higher-order terms)

$$x = x_c(\theta) + |\boldsymbol{p}|^2[3B(\theta)\cos\theta - B'(\theta)\sin\theta], \qquad (8)$$
$$y = y_c(\theta) + |\boldsymbol{p}|^2[3B(\theta)\sin\theta + B'(\theta)\cos\theta], \qquad (9)$$

where the curve $\{(x_c(\theta), y_c(\theta))\}_\theta$ is the facet contour corresponding to the zero-gradient limit of the curved region. Explicitly, we have

$$x_c(\theta) = \gamma(\theta)\cos\theta - \gamma'(\theta)\sin\theta, \qquad (10)$$
$$y_c(\theta) = \gamma(\theta)\sin\theta + \gamma'(\theta)\cos\theta. \qquad (11)$$

We should note that (10) and (11) are precisely the equations determining the *two-dimensional* ECS (=facet shape) from $\gamma(\theta)$ regarded as a one-dimensional interface tension. [13]

Let us now discuss the ECS near the facet edge in detail. For a given ECS described by an equation $z = z(x, y)$, we specify any point on the ECS surface by the two-dimensional position vector (x, y). For convenience, we take the xy plane (i.e., $z = 0$) to be the facet plane, which corresponds to putting $f(0) = 0$ in (4). Fix a point P on the facet contour, and choose the x and y axes so that the y-axis is parallel to the tangential line of the facet contour at P, and x-axis perpendicular to it (Fig.2). With this choice of the coordinate system, we have $\theta = 0$ at P and $\gamma'(0) = 0$ (see (5) and (11)). Our task is to obtain the ECS profile close to the point $P = (x_c(0), y_c(0)) = (x_c(0), 0)$.

Along the x-axis ($\theta = 0$, $\Delta x \equiv x - x_c(0)$), we have from (8) and (9)

$$|\boldsymbol{p}| = p_x = \frac{1}{\sqrt{3B(0)}}(\Delta x)^{1/2}, \qquad (12)$$

giving the "normal" profile with the well-known exponent $\theta_x = 3/2$:

$$z \sim \frac{2}{3\sqrt{3B(0)}}(\Delta x)^{3/2}. \qquad (13)$$

The profile (13) leads to the divergent behavior of the normal curvature $\kappa_x \approx \partial^2 z/\partial x^2 \sim (\Delta x)^{-1/2}$ near the facet edge. In the light of the universal *Gaussian* curvature jump at the facet edge, [6] the "tangential curvature" $\kappa_y \approx \partial^2 z/\partial y^2$, *along the x axis*, vanishes [6,14] as $(\Delta x)^{1/2}$ [Gaussian curvature is a product of two pricipal curvatures, κ_x and κ_y]. We should note that a different exponent θ_y for the tangential profile $z \sim (\Delta y)^{\theta_y}$ ($\Delta y = y - y_c(0)$) with $\theta_y > 2$ has already been implied by the vanishing of the tangential curvature at P.

The actual value of θ_y can be derived as follows. Note that, along the tangential line ($x - x_c(0) = 0$), θ and $|\boldsymbol{p}|$ are not independent but are constrained to satisfy,

$$-\frac{1}{2}\tilde{\gamma}(0)\theta^2 + 3B(0)|\boldsymbol{p}|^2 = 0, \qquad (14)$$

which can be derived by expanding (8) and (10) with respect to θ and $|\boldsymbol{p}|$ ($|\boldsymbol{p}| << 1$ and $|\theta| << 1$, very near the point P). Combining (14) with (8)–(11), we obtain

$$\theta = \frac{\Delta y}{\tilde{\gamma}(0)}, \qquad (15)$$

$$|\boldsymbol{p}| = \frac{\Delta y}{\sqrt{6\tilde{\gamma}(0)B(0)}}, \qquad (16)$$

along the y-direction. Near P, $-z = \tilde{f}(x_c(0), \Delta y + y_c(0))$ is expanded to give

$$-z = [\gamma(0) + \frac{1}{2}\gamma''(0)\theta^2]|\boldsymbol{p}| + B(0)|\boldsymbol{p}|^3$$
$$-\Delta y|\boldsymbol{p}|\theta - x_c(0)|\boldsymbol{p}|(1 - \frac{1}{2}\theta^2). \qquad (17)$$

Putting (15) and (16) into (17), we have

$$z = \frac{1}{3\sqrt{6B(0)\tilde{\gamma}(0)^3}}(\Delta y)^3, \qquad (18)$$

giving $\theta_y = 3$. We should remark here that the results (13) and (18) apply to any point on the facet contour, because the point P has been chosen arbitrarily; hence, $\tilde{\gamma}(0)$ and $B(0)$ in (13) and (18) can be replaced by $\tilde{\gamma}(\theta)$ and $B(\theta)$.

We have another simple geometrical derivation of (18) as follows. Very near the point P, we take a different point Q on the facet contour (Fig.3). Along the x'-direction chosen normal to the facet contour at Q, ECS

profile has the form (13) with $B(0)$ replaced by $B(\theta)$ where θ ($|\theta| \ll 1$) is the tangent angle (relative to y-axis at P) of the facet contour at Q. Note that the facet contour near P is approximately a part of a circle with its radius being the curvature radius R which is proportional to the step stiffness $\tilde{\gamma}(0)$. [15] For the normalized ECS we have $R = \tilde{\gamma}(0)$, hence, by an elementary geometry we can relate $\Delta x'$ (distance along the x'-direction) to Δy as

$$\Delta x' = \frac{(\Delta y)^2}{2R} = \frac{(\Delta y)^2}{2\tilde{\gamma}(0)}. \tag{19}$$

Replacing this $\Delta x'$ with Δx in (13) we reproduce (18) [we can put $B(\theta) \approx B(0)$, for our purpose].

We thus have shown that the "new exponent" $\theta_y = 3$ is a direct consequense of the well-established GMPT-type expansion of the vicinal surface free energy (4). Let us next discuss the "critical amplitudes" of the profiles. In the derivation of the universal Gaussian curvature jump at the facet edge, [6] the universal relation

$$B(\theta) = \frac{\pi^2 (k_{\mathrm{B}} T)^2}{6 \tilde{\gamma}(\theta)}, \tag{20}$$

which holds for any system (with only short-range inter-step interactions) is essential. This relation was originally derived in the coarse-grained TSK picture of the vicinal surface, [6] and has been confirmed in several ways. [6,7,8,16,17] For example, exact calculation for the BC-SOS model have verified (20) for arbitrary θ. [8] Using (20), we obtain a universal relation between the amplitudes of the normal and tangential profiles as follows. By $A_x(\theta)$ and $A_y(\theta)$ we denote the amplitudes of the ECS profiles, namely,

$$z \sim \begin{cases} A_x(\theta)(\Delta x)^{3/2} & \text{(normal direction),} \\ A_y(\theta)(\Delta y)^3 & \text{(tangential direction)} \end{cases}. \tag{21}$$

Restoring the scale factor λ in (3), we have from (13), (18) and (20),

$$[A_x^2(\theta) A_y(\theta)]^{1/3} = \lambda \frac{2}{3\pi k_{\mathrm{B}} T}, \tag{22}$$

which means that at a fixed temperature, the quantity $(A_x^2 A_y)^{1/3}$ is constant along the facet contour. The scale factor λ can be determined, for example, from the measurable ratio $\kappa/\sigma(\theta)^2$ ($\propto \lambda/(k_{\mathrm{B}} T)$ [15]) where κ is the curvature of the facet contour and σ the scaled fluctuation width of a single step. [18]

Before closing, we give a brief comment on the effect of the long-range inter-step interaction ($\sim 1/r^2$, with r being the ter-step distance) which has its origin mainly in the elastic deformation, and is important in discussing real crystal surfaces. [19] It has been known that inclusion of g/r^2 interaction with positive coupling constant g does not modify the GMPT-type form of expansion, but merely renormalizes the coefficient B. Explicit form of

the renomalized B has also been known. [16,20,21] Hence the exponent $\theta_y = 3$ does not change with the g/r^2 interaction. Since $\gamma(\theta)$ is a quantity associated with a single isolated step and is not affected by the g/r^2 interaction, the relation between γ and the renormalized B should be modified. If g is θ-independent (θ: mean running direction of steps) then the universal relation (22) still holds in a modified form. However, if g depends on θ, A_x and A_y will no longer be universally related to each other. In this case, (22) may provide a way to determine the θ-dependence of g experimentally, by measuring the ratio $[A_x^2(\theta) A_y(\theta)]^{1/3}/[A_x^2(0) A_y(0)]^{1/3}$.

To summarize, we have discussed the tangential profile of the equilibrium crystal shape near the facet edge below the roughening temperature. We have shown that the profile $z \sim (\Delta y)^{\theta_y}$ (Δy: distance along the tangential direction at the facet edge) with $\theta_y = 3$ is a direct consequence of the Gruber-Mullins-Pokrovsky-Talapov type expansion of the surface free energy $f(\boldsymbol{p}) = f(0) + \gamma(\theta)|\boldsymbol{p}| + B(\theta)|\boldsymbol{p}|^3 + \ldots$, which implies that the exponent 3 is universal. Further, we have presented a general relation between the amplitudes of the normal and the tangential profiles, which results from the known universal relation between the coefficient $B(\theta)$ and the step stiffness $\tilde{\gamma}(\theta) = \gamma(\theta) + \gamma''(\theta)$.

[1] E.E. Gruber and W.W. Mullins, J. Phys. Chem. Solids **28**, 6549 (1967); V.L. Pokrovsky and A.L. Talapov, Phys. Rev. Lett. **42**, 65 (1979).

[2] J. Villain and P. Bak, J. Phys. (Paris) **42**, 657 (1981); S.N. Coppersmith, D.S. Fisher, B.I. Halperin, P.A. Lee and W.F. Brinkman, Phys. Rev. Lett. **46**, 549 (1981); H.J. Schulz, B.I. Halperin and C.L. Henley, Phys. Rev. B**26**, 3979 (1982); T. Izuyama and Y. Akutsu, J. Phys. Soc. Jpn. **51**, 50 (1982); T. Izuyama and T. Yamamoto, J. Phys. Soc. Jpn. **52**, 4034 (1983).

[3] G. Wulff, Z. Kristallogr. **34**, 449 (1901); M. von Laue, Z. Kristallogr. **105**, 124 (1944).

[4] C. Jayaprakash, W.F. Saam and S. Teitel, Phys. Rev. Lett. **50**, 2017 (1983).

[5] C. Rottman, M. Wortis, J.C. Heyraud and J.J. Métois, Phys. Rev. Lett. **52**, 1009 (1984); Y. Carmi, S.G. Lipson and E. Polturak, Phys. Rev. B **36**, 1894 (1987); E. Rolley, E. Chevalier, C. Guthmann, and S. Balibar, Phys. Rev. Lett. **72**, 872 (1994).

[6] Y. Akutsu, N. Akutsu and T. Yamamoto, Phys. Rev. Lett. **61**, 424 (1988); T. Yamamamoto, Y. Akutsu and N. Akutsu, J. Phys. Soc. Jpn. **57**, 453 (1988).

[7] L.V. Mikheev and V.L. Pokrovsky, J. Phys. I (France) **1**,

373 (1991).

[8] R. Sato and Y. Akutsu, J. Phys. Soc. Jpn. **64**, 3593 (1995).

[9] J.D. Noh and D. Kim, Phys. Rev. E **53**,3225 (1996);*Extended universality of the surface curvature in equilibrium crystal shape*, cond-mat/9704177.

[10] S.R. Dahmen, B. Wehefritz and G. Albertini, *A novel exponent in the Equilibrium Shape of Crystals*, cond-mat/9802152.

[11] H. van Beijeren, Phys. Rev. Lett. **38**, 993 (1977).

[12] A.F. Andreev, Zh. Eksp. Theor. Fiz. **80**, 2042 (1981) [Sov. Phys. JETP **53**, 1063 (1982)].

[13] N. Akutsu and Y. Akutsu, J. Phys. Soc. Jpn. **56**, 2248 (1987); M. Holzer and M. Wortis, Phys. Rev. B **40**, 11044 (1989).

[14] P. Nozières and F. Gallet, J. Phys. (Paris) **48**, 353 (1987).

[15] M.P.A. Fisher, D.S. Fisher and J.D. Weeks, Phys. Rev. Lett. **48**, 368 (1982); Y. Akutsu and N. Akutsu, J. Phys. **A19**, 2813 (1986); J. de Coninck and J. Ruiz, J. Phys. A **21**, L147 (1988); D.B. Abraham and P.J. Upton, Phys. Rev. B **37**, 3835 (1988).

[16] T. Yamamoto, Y. Akutsu and N. Akutsu, J. Phys. Soc. Jpn. **63**, 915 (1994).

[17] T. Yamamoto, J. Phys. Soc. Jpn. **64**, 1945 (1995).

[18] N.C. Bartelt, R.M. Tromp, and E.D. Williams, Phys. Rev. Lett. **73**, 1656 (1994); N.C. Bartelt, J.L. Goldberg, T.L. Einstein, E.D. Williams, J.C. Heyraud and J.J. Metois, Phys. Rev. B **48**, 15453 (1993).

[19] K.H. Lau and W. Kohn, Surf. Sci. **65**, 607 (1977); V.I. Marchenko and A.Ya Parshin, Sov. Phys. JETP **52**, 52 (1981); N. Cabrela, J.M. Soler, J.J. Sáenz, N. García and R. Miranda, Physica **127B**, 175 (1984).

[20] W.F. Saam, Phys. Rev. Lett. **62**, 2636 (1989).

[21] N.C. Bartelt, T.L. Einstein and E.D. Williams, Surf. Sci. **276**, 308 (1992).

Figure Captions

Fig.1: Facet and vicinal surface. Upper right is an "atomic scale" view of the surface near the facet edge in the curved region, which can be regarded as an assembly of steps forming a vicinal surface.

Fig.2: Choice of x- and y-axes at P on the facet contour.

Fig.3: Geometrical derivation of Eq.(18). For Q very near P, "normal distance" $\Delta x'$ relates to "tangential distance" Δy as $\Delta x' = (\Delta y)^2/2R$, where R is the curvature radius at P.
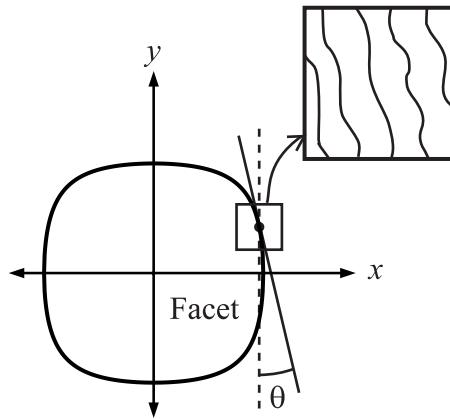
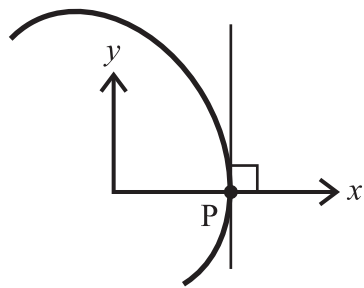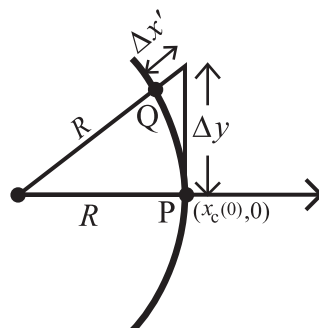Figure 1 Y. Akutsu, N. Akutsu and T. Yamamoto

Figure 2 Y. Akutsu, N. Akutsu and T. Yamamoto

Figure 3 Y. Akutsu, N. Akutsu and T. Yamamoto